# Contaminants of Emerging Concern and Wastewater Treatment


By

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For the

**Water Quality Program**

Washington State Department of Ecology

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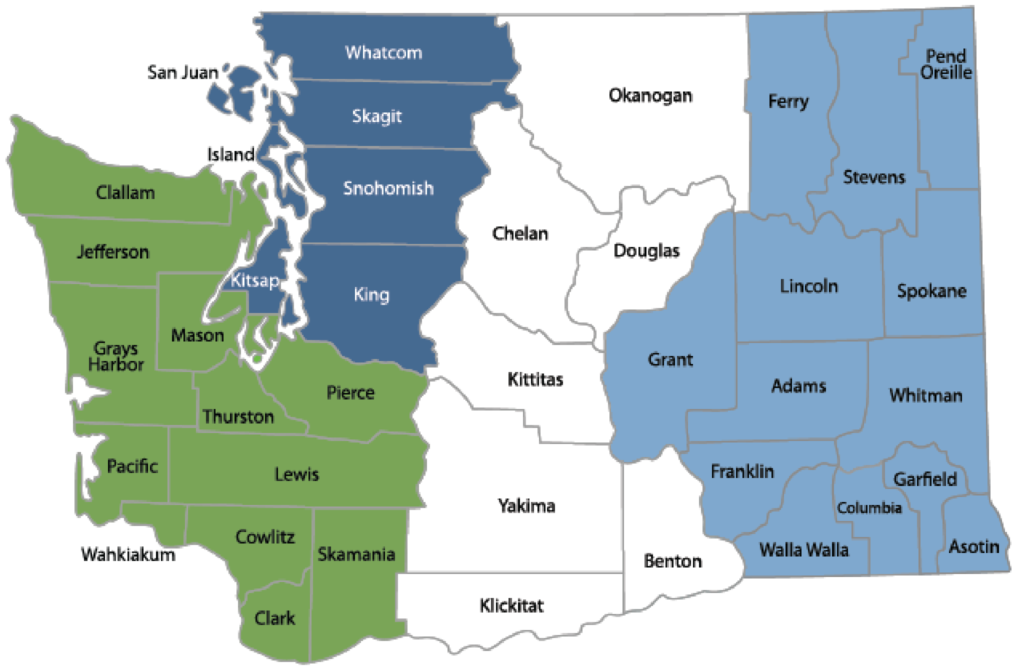


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DEPARTMENT OF
ECOLOGY
State of Washington

# Table of Contents

# List of Figures and Tables

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# Executive Summary

Contaminants of Emerging Concern (CECs) are contaminants, both natural and synthetic, that may cause ecological or human health effects and are not widely regulated. CECs found in wastewater include, but are not limited to, pharmaceuticals, personal care products (e.g., synthetic fragrances, antibacterial compounds), plasticizers, food additives, flame retardants, microparticles, and per and polyflouryl alkyl substances (PFAS).

These CECs can end up in wastewater through common activities like washing clothes, flushing the toilet, bathing, and maintaining and cleaning a home or business. CECs also reach wastewater treatment plants (WWTPs) from industrial processes, hospital waste, and recreational activities or settings like parks (i.e., boat washing and rinse off stations).

Many WWTP technologies exist that have the ability to remove some or all CECs from wastewater. Adding more advanced treatment to a WWTP can reduce CECs, with the removal rate dependent upon the approach chosen and other implementation factors.

The purpose of this paper is to explore new and traditional treatment options for wastewater treatment plants (WWTP), highlight the potential co-benefits some of these technologies may bring, and provide information on options for reducing CECs in wastewater.

This paper outlines the treatment effectiveness of fifteen treatment technology types by comparing the percent removal for four common CECs (caffeine, carbamazepine, triclosan, and ibuprofen), potential nutrient removal co-benefits, and advantages and disadvantages for consideration (see Appendix B).

Carbon filtration, ozonation, and reverse osmosis were the most successful CEC removal technologies across these four CECs. Among these three technologies, only reverse osmosis successfully removes nutrients. However, WWTPs often combine technologies to remove multiple contaminants across a treatment train.

Among technologies directly capable of nutrient removal with the right configuration, the most CEC removal is achieved by MBBRs (an attached growth system), followed by MBRs, BNR, and other attached growth systems. Conventional activated sludge, constructed wetlands, and onsite systems also remove CECs, but at lower rates. This paper found that microfiltration and ultrafiltration alone will <u>not</u> likely be successful approaches for removing CECs. Nanofiltration will have mixed success, and biosorbant removal rates are highly dependent on the product used. However, results can still be highly variable due to local conditions.

WWTP managers should take into account that operational changes such as increasing the sludge retention time or the hydraulic retention time, or both, to reduce nutrient effluent concentration also allows for more time for CECs to be reduced via biodegradation and/or adsorption. Compound removal efficacy will change as a result of variations in operational procedures, environmental conditions, and WWTP influent characteristics.

For most CECs, concentrations may be reduced but the chemicals will not be entirely removed from WWTP effluent and the pollutant loadings to the environment could remain substantial. Some CECs are so recalcitrant that they require advanced treatment for removal. Not all CECs can be removed with nutrient removal technologies. More advanced treatment like advanced oxidation plus filtration may be needed to remove the majority of CECs from wastewater. Non-treatment approaches including source control, product replacement, and education, and outreach campaigns to encourage proper disposal of pharmaceuticals are necessary to reduce the overall amount of CECs discharged in wastewater from WWTPs.

# Introduction

Contaminants of Emerging Concern (CECs) have been found in Washington's waters across the state (Meador et al., 2016; Morace, 2012; Partnership, 2007; Tian et al., 2019). CECs are contaminants, both natural and synthetic, that may cause ecological or human health effects and are not widely regulated. There are currently an estimated 25,000 - 84,000 chemicals used in commerce today and new potential CECs are added to the market every year (IOM, 2014). Examples of CEC categories typically found in wastewater include pharmaceuticals, personal care products, food additives, and industrial and commercial compounds.

The purpose of this paper is to explore new and traditional treatment options for wastewater treatment plants (WWTP), highlight the potential co-benefits some of these technologies may bring, and inform options for reducing CECs in wastewater discharges. Understanding the removal mechanisms and rates of each WWTP technology will allow for more informed decision-making for policy makers and wastewater treatment plant designers and operators.

## Sources of Control and Wastewater CES's

CECs in municipal wastewater from **domestic and residential sources** are generated by seemingly innocuous everyday tasks like:

- **Washing clothes** – Microplastics from synthetic materials can break away and enter the environment. Per- and polyfluoroalkyl substances (PFAS) can also enter wastewater from waterproof and stain resistant clothing. Other personal care products, like sunscreens and lotions, can wash off from clothing as well.
- **Flushing the toilet** – A lot of pharmaceuticals enter the environment through liquid and solid human and animal waste. Pharmaceuticals or their metabolites pass through living beings and are excreted.
- **Bathing** – Personal care products can contain CECs and wash off while bathing. Examples include moisturizers, make up, shampoos, conditioners, and hair dyes.
- **Household Cleaning and Maintenance** – Some cleaning products and household maintenance supplies contain CECs. This includes antimicrobial cleaning products, tile cleaner, glues, household pesticides, etc.

CECs also come from industrial sources, hospitals, and other facility-specific sources of wastewater:

- **Industrial waste** – Some WWTPs receive water from industrial sources. These sources can contain CECs. Many of these industrial sources are treated prior to entering a WWTP, known as pretreatment. However, developing pretreatment programs and standards for specific pollutants is a long and involved process. Therefore pretreatment programs are not nimble enough to address all CECs in industrial wastewater, and CECs end up in WWTPs.

- **Medical facilities' wastewater** – Wastewater from hospitals, nursing homes, and other medical facilities often contain more pharmaceuticals than household wastewater due to the increased use of pharmaceuticals in the population. The type of pharmaceuticals entering waste can be more toxic (e.g., chemotherapies, contrast used for medical imaging). In some cases, hospitals have a pretreatment program in place to reduce the effects of the pharmaceutical-laden wastewater on a WWTP. However, no hospitals in Washington are regulated by a local or state pretreatment program that addresses pharmaceuticals and other CECs.
- **Other wastewater** – Other wastewaters include "washwaters" from recreational spray parks and outdoor washing of recreational equipment that can contain personal care products, waxes, microplastics, antifouling agents, etc.
- **Runoff** – stormwater can collect and convey CECs when rain falls on a contaminated surface. Runoff can enter wastewater treatment plants through infiltration and inflow. Although beyond the scope of the paper, CECs in stormwater were included as a source and as a research recommendation in the recommendation section due to recent research that shows that runoff can contain CECs.

There are more than 2.8 million households in Washington State that produce wastewater. This number does not include industrial sectors, office buildings, and other commercial areas that are connected to wastewater treatment facilities and can contribute CECs other than pharmaceutical and personal care products. Treatment at the source is not always feasible, and complete control and mitigation of CECs is challenging.

Everyone who uses pharmaceuticals has the potential to be a CEC source because some of the compounds are not completely absorbed by the body and pharmaceutical metabolites will still be excreted. Pharmaceuticals have not been restricted or controlled for water quality purposes. This means that pre-treatment and WWTPs are at the front line for removing many CECs from wastewater.

Source controls such as product substitutions for less toxic chemicals or unused pharmaceutical take-back programs already exist, but these efforts are generally limited in scope and effectiveness, and do not offer a full solution to prevent CECs from entering wastewater.

## Effects of Wastewater CECs on Aquatic Life

CEC susceptibilities vary with species. Recent research on CECs in wastewater has demonstrated biological impacts such as: negative metabolic changes in Chinook salmon (Meador et al., 2016), endocrine disruption in multiple fish species (Brodin et al., 2014; Harding et al., 2006), reduced fertility in fathead minnows (Niemuth & Klaper, 2015), increased antibiotic resistant bacteria, general increased morbidity in Coho and Chinook salmon (Meador, 2014), and bioaccumulation in annelids (Kinney et al., 2015).

Some organisms (e.g. birds and fish) may have negative biological impacts because they do not have the ability to successfully process and excrete some CECs that are made for other target species (e.g., mammals like humans and livestock) (Fent et al., 2006).

CECs may have low acute toxicity but have significant chronic or sub-lethal effects (e.g., reproduction, growth, development, etc.) at low levels of exposure. Additionally, full effects of exposure may not develop until later in a species' lifespan. According to EPA guidance: "Traditional toxicity endpoints may not be sufficiently comprehensive for criteria derivation for these chemicals and the chemicals may also have specific modes of action that may affect only certain types of aquatic animals (e.g., vertebrates such as fish)" (EPA, 2008). Little is also known about aggregate toxic effects of multiple CECs on aquatic species. Toxic effects of a CEC can be compounded by other CECs present which is often the case with wastewater.

Later sections of this paper focus on four representative CEC chemicals: triclosan, caffeine, ibuprofen, and carbamazepine. The full effect of these four chemicals on aquatic biota are not fully understood. As mentioned above, traditional toxicity endpoints for CECs do not always represent how toxic a CEC is on species. However, Olaniyan et al., 2016 found that triclosan is a carcinogen for multiple aquatic species. Triclosan is toxic to algae (Tatarazako et al., 2004). Caffeine and ibuprofen can be toxic, but not at concentrations found in wastewater. Lastly, carbamazepine is lethal to many aquatic insects (Oetken et al., 2005).

# Regulation of CECs

Clean Water Act (CWA) National Pollutant Discharge Elimination System (NPDES) permits regulate WWTP discharges to surface waters and a State Waste Discharge (SWD) Permit for WWTP discharges to groundwater. However, CECs in wastewater can be difficult to regulate through discharge permits. CECs do not yet have water quality criteria in EPA-approved water quality standards because of the challenges associated with use of traditional toxicity endpoints discussed above. This generally precludes permit writers from assigning water quality-based effluent limitations in permits. Also, insufficient data on CEC control and treatment efficacy generally precludes the derivation of technology-based effluent limits in permits. Furthermore, the list of CEC compounds is ever changing and expanding – as we learn more about a compound, permitting authorities may be able to control it more like a traditional pollutant, and as we use more sophisticated analytical methods to test what is in our consumer products, water, sediment, and aquatic life, it is likely that more and more compounds will become an "emerging concern".

CEC compounds and concentrations are variable over space and time with little chronic or acute ecological toxicology information for the CECs or their degradants. For example, WWTPs that serve a community with an older population might have higher concentrations with a more variable matrix of pharmaceuticals and personal care products (PPCPs). Other WWTPs might receive wastewater from industry or hospitals that would create a different influent mix of CECs.

CEC occurrence can fluctuate temporarily due to allergies and illness that vary over seasons (e.g., allergy medications, influenza medications, sunscreen, DEET, etc.) or due to increased use in antimicrobials and cleaning products during widespread illness outbreaks (e.g., Covid-19 pandemic or local flu outbreaks). While highly effective in controlling many pollutants, the NPDES permitting process requires a lot of monitoring and ecotoxicity information and, therefore, is currently not well-suited to address many CECs. There are over 40,000 chemicals used in commerce. Each would require an approved analytical method and ecotoxicity data to promulgate water quality criteria and derive effluent limits in wastewater discharge permits, and this is currently not possible with existing resource limitations.

# Case Studies and Regulation

Switzerland, the Netherlands, Sweden, Germany, Denmark, and Belgium have all either started to explore or to implement end of pipe technologies at WWTPs to limit the amount of micropollutants (CECs) in effluents (EurEau, 2019). These countries are modeling their efforts off of Switzerland's required WWTP upgrades, also known as The Swiss Strategy (Eggen et al., 2014).

Switzerland saw a marked reduction in fish population due to ecotoxicity from WWTP discharges. In 2009, this led them to begin a twenty-year initiative to upgrade their WWTPs. In 2011, Switzerland revised their Water Protection Ordinance to require additional treatment for CECs. The Swiss Federal Office of the Environment (FOEN) settled on ozonation plus granular activated carbon (GAC) at WWTPs because the combined technologies removed 80% of all CECs found in their wastewater during lab tests (Eggen et al., 2014). FOEN highlights the need for source control and a public education campaign about proper disposal of pharmaceuticals and chemicals. However, they also recognize that ongoing use of some CECs is unavoidable, like pharmaceuticals, and that others will take time to phase out through green chemistry and behavior modifications.

FOEN's ordinance requires additional treatment for all WWTPs serving more than 80,000 people, all WWTPs serving more than 24,000 people and discharging into lakes, and all WWTPs serving more than 8,000 people and discharging into rivers, if the discharge represents more than 10% of the minimum flow. Based on these thresholds, 100 out of 700 WWTPs in Switzerland will need to upgrade their systems by 2035 (Joss et al., 2016). This ordinance is financially feasible because it does not require upgrades for all treatment plants and only focuses on the treatment plants with the most impact to local aquatic health.

Sweden began the same process in 2014, and is requiring the installation of ozonation followed by GAC to address CECs in wastewater in some WWTPs. The Swedish Agency for Marine and Water Management (SWAM) found altered fish behavior in the Baltic Sea due to WWTP CEC loading. To date they have funded projects for eight wastewater treatment plants to install enhanced treatment.

All eight plants had successful removal rates from a broad range of CECs. SWAM is continuing to fund research and upgrades for smaller WWTPs across Sweden (Cimbritz & Mattsson, 2018).

In 2018, the German Environmental Agency (GEA) produced a set of recommendations for the reduction of CECs in wastewater. They recommend installing a fourth stage of treatment for WWTPs. However, they did not outline what the fourth stage would entail (Ahting et al., 2017).

# Previous Studies and Initiatives in Washington

Ecology has participated in and led multiple studies regarding CECs over the last 15 years.

In 2008, Ecology conducted a study to investigate per- and polyfluoroalkyl substances (PFAS), a CEC of nationwide interest, in 14 surface waters and in the effluent from four WWTPs. The study found that every WWTP effluent sample contained PFAS at higher concentrations (61 – 418 ng/L) than the surface waters (~10 ng/L) (Furl & Meredith, 2010). This study gave Ecology a baseline of PFAS data for another study in 2016 (Mathieu & McCall, 2017) that showed lower, but not significantly lower, concentrations in WWTP effluent. Additionally, the study found changes in the composition of PFAS species and made recommendations for further studies to more fully characterize the PFAS concentrations around the state.

Previously, Ecology partnered with the Environmental Protection Agency (EPA) and the Puget Sound Partnership as part of a National Estuary Program (NEP) funded project that aimed to estimate sources of toxic chemicals and potentially identify ways to restore the environmental health of Puget Sound by 2020 (Ecology et al., 2007). The main goals were to:

- Identify toxic chemicals that have harmed or threaten to harm the Puget Sound ecosystem or the beneficial uses which humans obtain from the Sound.
- Estimate the loading rates of key contaminants from their sources through their major pathways to Puget Sound.
- Provide information that will support development of a strategy to identify the actions, practices, and policies necessary to protect and restore the overall health of the Puget Sound ecosystem.

Ecology broke this effort down into three phases: Initial Estimation of Toxic Chemical Loadings to Puget Sound, Improving the Loading Estimate, and Targeting Priority Toxic Sources. Ecology published two Phase 3 studies in 2010. The first concluded that wastewater is a significant source of CECs. The second, entitled “Phase 3: Pharmaceuticals and Personal Care Products in Municipal Wastewater and Their Removal by Nutrient Treatment Technologies” (Lubliner et al., 2010), assessed the efficacy of nutrient removal technologies on removing CECs and PPCPs, and serves as the foundation for this literature review and findings report. Findings of this 2010 report included:

- PPCPs were routinely detected in municipal wastewater.
- Some of the PPCPs that were removed from the wastewater were subsequently found in the biosolids.
- Enhanced biological nutrient removal for phosphorus and nitrogen, and chemical addition with filtration for phosphorus removal removed 31 more PPCP analytes from the wastewater.
- Three PPCPs (carbamazepine, fluoxetine, and thiabendazole) were left relatively untreated by these treatment technologies.

In 2019, the Southern Resident Killer Whale (SRKW, also known as Orca) Task Force identified toxics contaminants in the environment as one of the three main threats facing orcas in Puget Sound. The SRKW Task Force Recommendation Report included two recommendations to address toxics harmful to orcas and their prey:

1. Recommendation 30: Identify, prioritize and take action on chemicals that impact orcas and their prey.
    - By March 2019, the Department of Ecology should develop a prioritized list of chemicals of emerging concern that threaten the health of orcas and their prey and pursue policy and/or budget requests in the 2019 legislative session to prevent the use and release of chemicals of emerging concern into Puget Sound.
    - Direct Ecology to convene discussions and develop a plan to address pharmaceuticals, identifying priorities, and source control and wastewater treatment methods.
    - Periodically review and update toxicological information as new science emerges and adaptively manage plans and programs.
2. Recommendation 32: Improve effectiveness, implementation, and enforcement of National Pollutant Discharge Elimination System permits to address direct threats to Southern Resident orcas and their prey.
    - Update aquatic life water quality standards focused on pollutants most harmful to Southern Resident orcas and their prey.
    - Direct the Department of Ecology to consider developing stronger pre-treatment standards for municipal and industrial wastewater discharges under NPDES.
    - Provide funding for Ecology to increase inspections, assistance programs, and enforcement to achieve water quality standards. Prioritize enforcement where limits are exceeded for pollutants known to be harmful to Southern Resident orcas.

Ecology is currently working through these SRKW Task Force recommendations. This paper serves as the foundation for many of the recommendations and sub-bullets listed.

Ecology is currently developing a NPDES General Permit to address nutrient discharges from WWTPs that are causing or contributing to water quality problems (low dissolved oxygen conditions (A. Ahmed et al., 2019)) in the greater Puget Sound area. This general permit is expected to result in upgraded technology and operations at many WWTPs to achieve nutrient removal in the years ahead. Some of the advanced technologies to remove nutrients will also remove CECs (Lubliner et al., 2010).

# Treatment Processes

CECs are removed through commonly used treatment processes: sorption, biodegradation, membrane separation, oxidation, and volatilization. Treatment technologies are designed to optimize these basic processes. Below is a brief explanation of how each process works:

## Sorption

Sorption includes two distinct processes; adsorption and absorption. Adsorption is the most prevalent removal mechanism of CECs. It occurs when CECs adhere to a surface of a solid via ionic, covalent, or metallic bond.

Adsorption rate is affected by many different environmental factors and chemical characteristics of the CECs. Environmental factors include pH, temperature, and retention time. Chemical characteristics include hydrophobicity, acid disassociation constant, chemical structure, and surface area of adsorbent.

Adsorption due to ionic bonds is weak and the CEC can quickly desorb due to a change in the environment (pH, temperature, etc.). Covalent bond adsorption results in a stronger bond that is less likely to break. Figure 1 shows adsorption via covalent and ionic bonds and absorption.

Absorption is a less commonly used treatment process than adsorption. It occurs when a CEC molecule permeates into a solid as shown in Figure 1.

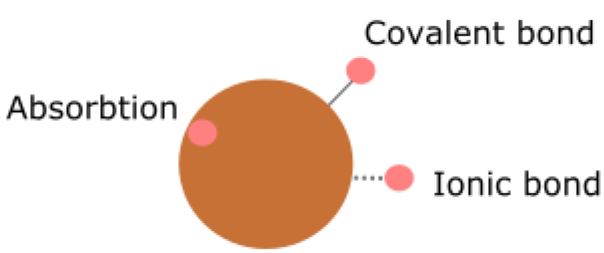


**Figure 1. Absorption and Two Types of Adsorption**

## Biodegradation and Biosorption

Biodegradation is the decomposition of material through microorganisms like bacteria and fungi. This is a very common wastewater treatment process. Biological removal of nutrients from wastewater relies on microorganisms. However, many of the microorganisms used and cultivated for nutrient removal are not optimized to break down complex CECs. Often wastewater contains more energetically favorable carbon sources that microorganisms preferentially consume. Additionally, a lot of CECs are antimicrobial in nature or can be toxic to microorganisms. Like all removal mechanisms, biodegradation is highly variable (Tran et al., 2013). Chemical structure, light, water, oxygen, temperature, redox potential, and types of organic material can all effect removal rates of CECs via biodegradation. Figure 2 demonstrates how different microbes can break down compounds into smaller and smaller constituents.

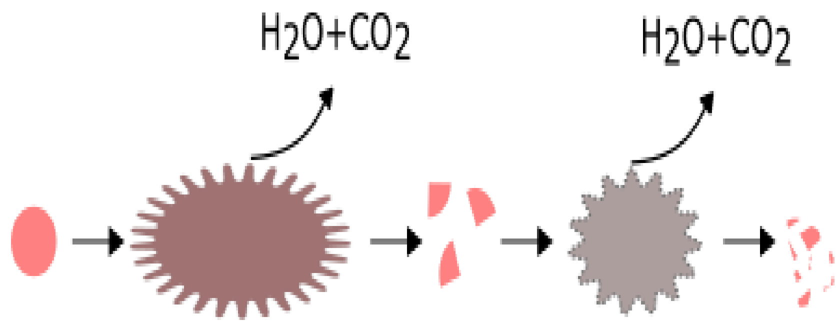


Figure 2. Aerobic Biodegradation

## Membrane Separation

Membrane separation occurs when a membrane or filter separates material via pores in the structure of the membrane. These pores come in different sizes as shown in Figure 3: Microfiltration (MF), Ultrafiltration (UF), Nanofiltration (NF), and Reverse Osmosis (RO). RO requires a filter with the smallest pore size and the highest amount of pressure (0.0001 µm), and microfiltration requires the lowest amount of pressure and the largest pore size (0.1 µm). RO is able to filter out inorganic ions whereas microfiltration can only filter out larger materials like parasites, bacterium and coagulated suspended solids. Microfiltration and ultrafiltration are unlikely to remove CECs unless they have adsorbed onto or absorbed into larger particles (Radjenovic et al., 2007).

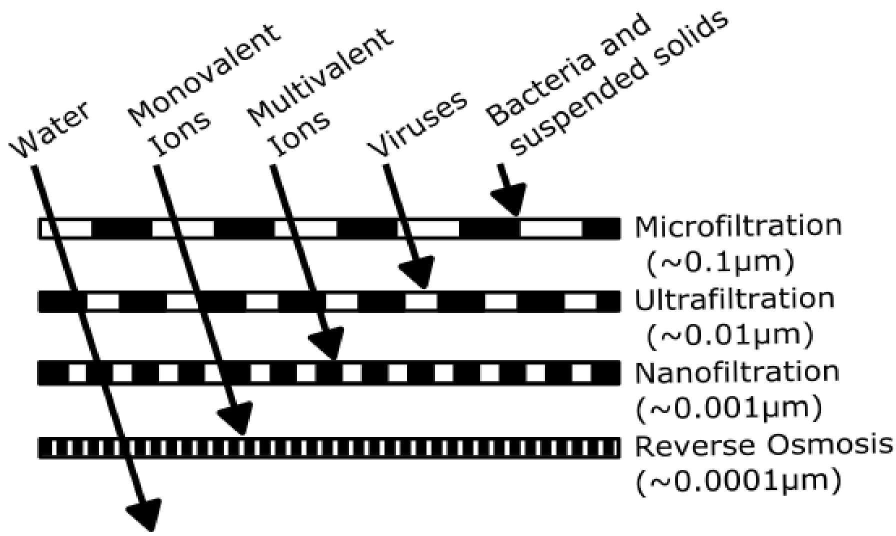


**Figure 3. Pore Size and Rejection of Different Filters**

## Oxidation

Chemical oxidation is a process by which electrons are transferred from an oxidizing reagent to a chemical species, thus breaking down the chemical. Hydroxyl radicals ($OH^-$) are the main oxidant when dealing with an aqueous solution. Hydroxyl radicals are generated with a catalyst like UV light or by infusing ozone into the solution (see Figure 4). Oxidants are able to break down CECs into byproducts. Efficacy and efficiency of oxidation depends on the CECs in the wastewater, the oxidant used, and time. Oxidation is often followed by filtration in order to remove potentially harmful byproducts.

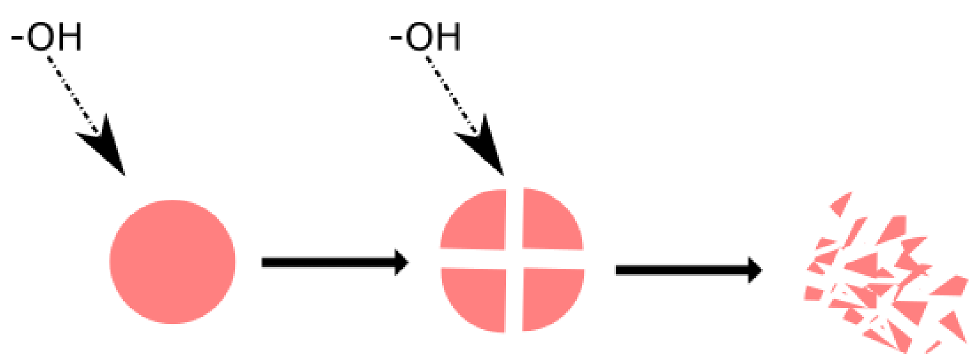


**Figure 4. Breakdown of a Molecule via Oxidation**

## Volatilization

Volatilization occurs when a dissolved CEC vaporizes and enters the atmosphere. Volatilization does not necessarily remove a CEC from the environment, rather it transfers the CEC from the wastewater into another phase. CEC removal by volatilization is negligible (Jones et al., 2005) and so, for the purposes of this paper, volatilization will not be discussed.

# Wastewater Treatment Process in Washington

Washington State NPDES regulations currently require a minimum level of secondary treatment and effluent requirements to protect human health and aquatic life from domestic wastewater discharges to surface waters (WAC 173-221). Municipal WWTPs across Washington State utilize a wide array of approaches to manage domestic sewage to meet NPDES permit requirements and the needs of their community. Treatment systems range from small, passive systems to large advanced treatment systems at WWTPs.

Rural or small communities often do not have centralized wastewater treatment systems. Centralized systems are impractical in these areas because of the smaller amounts of wastewater created, the distance to a centralized location, or terrain that makes it difficult to transport waste. These communities treat their wastewater closer to the source and rely on smaller, decentralized systems that are not complicated to operate.

WWTPs often use a combination of primary and secondary (and occasionally tertiary/advanced) treatment systems. This treatment train approach takes advantage of using multiple technologies installed in series. Figure 5 shows the flow-through of a treatment train and multiple treatment technologies. Each treatment technology has different removal mechanisms and removal rates for CECs. The makeup of a treatment train depends on the permit requirements that a WWTP needs to meet. Currently, only one WWTP discharging to the Puget Sound includes advanced treatment in their treatment train for nutrient removal. Three municipal treatment plants discharging to the Spokane River utilize different configurations of advanced treatment to meet water quality based effluent limits stemming from an EPA approved dissolved oxygen total maximum daily load (TMDL). There are also other plants around the state that utilize enhanced secondary treatment; however, the majority of WWTPs in Washington State use conventional secondary treatment technologies.

Primary treatment consists of screening, grit removal, and primary clarification. These steps do not remove many CECs from wastewater with the notable exception being larger particles, like plastic debris, that the grit screen or the settling tank can remove. Some CECs may be bound to these plastic particles, but likely not in meaningful quantities.

As regulatory requirements evolve, WWTPs may incorporate new tertiary/advanced treatment technologies. Adding multiple tertiary and advanced treatments can greatly reduce CECs in the final effluent. However, complex treatment trains also increase the need for increased physical footprint for treatment, energy, and operational expertise; all of which tend to increase both the initial costs and long-term operational costs of wastewater treatment.

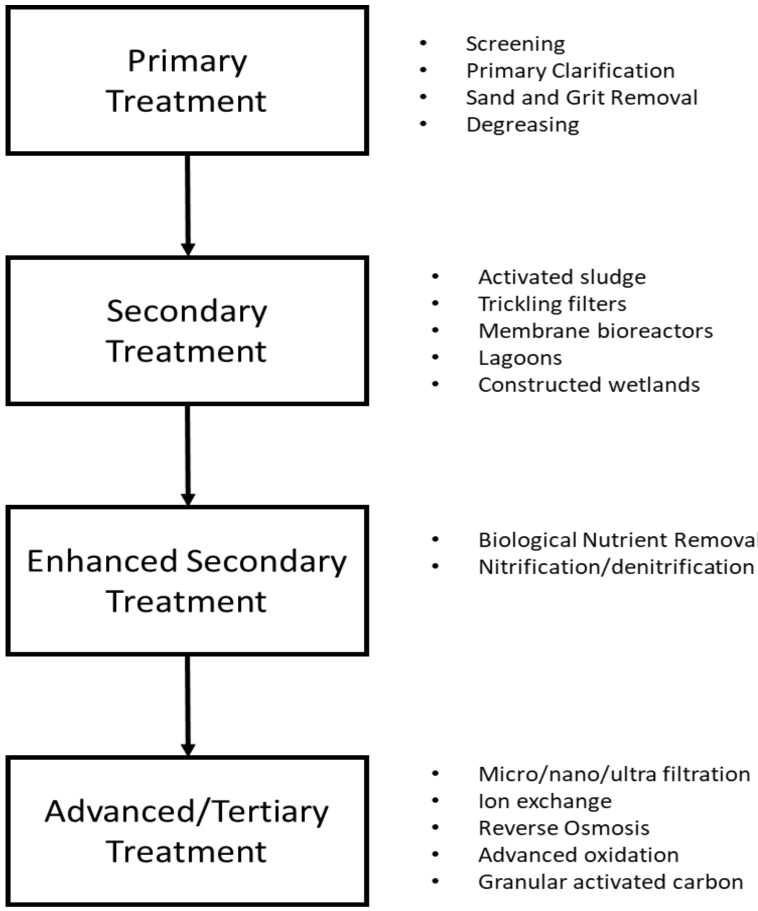


Figure 5. Outline of a treatment train and potential technology options

All treatment systems reduce the concentrations some CECs, but none remove all CECs. There is no “one size fits all” approach to CEC removal by wastewater treatment because CECs have very different chemical characteristics and there are so many CECs in common use that there is often insufficient data on occurrence and concentration of these chemicals in the wastewater. Additionally, the treatment technologies that are available can have highly variable removal rates for the same compound depending on the season, the day (Vieno et al., 2005), location of the WWTP, or the chemical make-up of the influent. Some compounds are recalcitrant and can only be removed by complex and expensive advanced/tertiary treatment technology like granular activated carbon, reverse osmosis, and ozonation.

# Wastewater Treatment Technologies

Below is a list of treatment technologies and a summary of the benefits, drawbacks, and published removal rates of four common and representative CECs in wastewater. The following CECs were chosen because they are well studied, which means most of the technologies addressed in this paper had published removal results. Additionally, these CECs are ubiquitous in wastewater and span the range from easy to remove (caffeine, ibuprofen) to hard to remove (triclosan) to extremely recalcitrant (carbamazepine):

- **Caffeine** is a naturally occurring chemical used as a stimulant of the brain and central nervous system. Caffeine is often used in WWTP technology efficacy studies because it is ubiquitous in WWTPs.
- **Ibuprofen** is a nonsteroidal anti-inflammatory drug that works by reducing hormones that cause inflammation and is used to reduce fever and pain.
- **Triclosan** is an antibacterial and antifungal agent that was added to many consumer personal care products such as toothpaste, antibacterial soaps, and body washes. The FDA banned the use of Triclosan in 2016 in consumer products, but it can still be found clothing, kitchenware, toys and as a pesticide.
- **Carbamazepine** is an anticonvulsant/analgesic drug used to treat seizures, mania, and neuropathy. It is often used in WWTP technology efficacy studies because it is well known to be extremely recalcitrant.

## Onsite Sewage Systems (Septic Systems)

Well-designed and properly maintained on-site sewage systems function similarly to post-secondary wastewater treatment processes (Schaider et al., 2017). The majority of CEC removal in septic systems occurs in the drainfield through sorption and biodegradation rather than in anaerobic digestion in the septic tank itself (Conn et al., 2006). CECs that are hydrophobic adsorb to the soil more efficiently after entering the drainfield and are therefore removed more easily. Generally, on-site sewage systems do not effectively remove hydrophilic CECs (Schaider et al., 2017) and therefore can be considered pathways for these CECs to reach groundwater and in some cases, eventually surface water.

Onsite sewage systems with additional aerobic treatment steps have removal rates of CECs that are comparable to conventional secondary WWTP technologies (Conn et al., 2006; Du et al., 2014; Yang et al., 2016, Garcia et al., 2013). The additional aerobic step consists of either a system in which wastewater is sent through aerobic textile-media biofilters, or one that has a two-tank system followed by a subsurface flow constructed wetland. However, these more advanced systems are not the norm for residences connected to septic systems.

Schaider et al., 2017 conducted a review of organic wastewater compound concentrations and removal in onsite wastewater treatment systems. They found that wastewater entering on-site sewage systems can be highly variable because they typically come from residential households, whose personal practices and use of CECs often differ (e.g. different pharmaceuticals taken, different cleaning supplies and personal care products used, and different disposal habits). Furthermore, they observed a lot of variation between laboratory and field results, where unpredictable environmental factors can influence removal rates. On-site sewage systems and associated drainfields are highly susceptible to changing outdoor conditions, like temperature, which greatly alter removal rates making it difficult to predict CEC removal rates in onsite sewage treatment systems and even harder to determine which onsite systems are a significant source of CECs.

Advantages and disadvantages of onsite sewage systems as a CEC and nutrient treatment technology are highlighted in Table 1.

**Table 1. Onsite Sewage Systems advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[2] |
|---|---|---|---|
| • CEC removal rates on par with conventional activated sludge<br>• Good for remote areas without access to larger facilities | • Does not remove recalcitrant compounds<br>• Not easily upgraded<br>• Removal rates are highly variable based local conditions<br>• Can fail | • Some nutrient removal does occur<br>• Can also be a source for nutrients when drainfield fails or is poorly designed<br>• Hard to determine when a drainfield will fail | Caffeine- 99.63%<br>Ibuprofen- 76%<br>Triclosan- 80%<br>Carbamazepine- 8% |

## Constructed Wetlands

Constructed wetlands can be a cost effective treatment option for small capacity systems where land price and space availability are not factors. The main types of constructed wetlands for wastewater treatment are: free water surface (FWS), and Subsurface Flow Wetlands (SFW). FWS resemble natural wetlands with pockets of vegetation and standing water. SFW are smaller with gravel beds that might have vegetation planted overtop. They are often used in conjunction with septic tanks and can replace a drainfield. Constructed wetlands have both advantages and disadvantages in nutrient and CEC removal which are highlighted in Table 2.

[2] (Schaider et al., 2017)

Ávila, Nivala, et al., 2014 found that, similar to on-site sewage systems, constructed wetlands that contain a hybrid vertical and horizontal flow system, or have multiple constructed wetlands in series were more efficient at removing CECs. The average overall removal efficiency of all studied CECs[3] was high (87% removal efficiency, ± 10%) with the antibiotics (43% removal efficiency, ± 32%) as an exception. Notably, this study only included three of the nine highest priority chemicals as defined by De Voogt et al., 2009. Hybrid systems allow for wastewater to move through variable conditions with longer attenuation which can promote different removal pathways.

**Table 2. Constructed Wetlands advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[4] |
|---|---|---|---|
| • Long HRT which increases removal rates<br>• More mechanisms for removal than on-site sewage systems<br>• Good for remote areas without access to larger facilities | • Requires space<br>• Variable CEC removal rates<br>• Does not remove recalcitrant CECs<br>• Susceptible to changing environmental conditions<br>• Requires large land area which can increase the cost | • Higher nutrient removal rates than lagoons and on-site sewage systems on average | Caffeine- 80.3%<br>Ibuprofen- 80%<br>Triclosan- 58-79%<br>Carbamazepine- 26.7% |

## Lagoons

Lagoons are often used by rural municipalities because they are easy to maintain and have low operational costs. Lagoons act similarly to constructed wetlands. Lagoons remove CECs through multiple methods (biodegradation, sorption, and some UV radiation if exposed to sunlight). Therefore, lagoons can remove CECs that easily sorb or biodegrade. Lagoons also have a wide variation in removal rates because there are different exposures to environmental elements and differences in construction. Therefore it is hard to model how effective lagoons are at removing CECs (Hoque et al., 2014; X. Li et al., 2013).

However, studies show that CECs are removed at similar rates by lagoons and by WWTPs with secondary treatment technologies installed. Hoque et al., 2014 found that Lagoons removed 0-40% of carbamazepine, 78-91% of ibuprofen, and 42-97% of triclosan present in the influent with the variation in removal rate due to seasonal changes.

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[3] Avila, Nivala, et al, 2014 study looked at the following CECs: Enrofloxacin, sulfamethoxazole, doxycycline, erythromycin, lincomycin, ibuprofen, diclofenac, tonalide, oxybenzone, triclosan, bisphenol A, ethinylestradiol

[4] (Arous et al., 2019; Auvinen et al., 2017; Ávila, Bayona, et al., 2014; Zhang et al., 2011)

Li et al., 2013 found similar results with all pharmaceutical concentrations, except carbamazepine, reduced by 88-100% in warmer months. Advantages and disadvantages of lagoons as a CEC and nutrient treatment technology are highlighted in Table 3.

**Table 3. Lagoons advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates [5] |
|---|---|---|---|
| • Long retention time which increases removal rates<br>• Inexpensive and easily maintained<br>• Good for remote areas without access to larger facilities | • Does not treat recalcitrant CECs that do not easily adsorb<br>• Variable removal rates depending on local conditions<br>• Requires large land area, which can increase the cost | • Often unable to meet ambient aquatic life criteria for ammonia | Caffeine- 100%<br>Ibuprofen- 91.5%<br>Triclosan- 97.2%<br>Carbamazepine- <0% |

## Conventional Activated Sludge (CAS)

This process mainly relies on biodegradation and adsorption for the removal of CECs. Microorganisms in the wastewater break down CECs into byproducts. Given sufficient time and the right conditions (redox potential, temperature, etc.), the microorganisms can break down the CECs and their byproducts all the way to water and carbon dioxide. However, CEC influent concentrations are very low (nanograms to micrograms per liter) and cannot support rapid colonization of microorganisms that specialize in breakdown of CECs. Clara et al., 2005 found that biodegradable CECs have a corresponding sludge retention time (SRT) critical value, wherein any facility with a lower SRT will not effectively remove the corresponding CEC. SRT is the mean time that the sludge remains in the system. The sludge contains the microorganisms needed to remove CECs, and thus SRT can also describe the regeneration time frame of the microorganisms. Ibuprofen has an SRT critical value of about 5 days. Bezafibrate, a more recalcitrant PPCP, has a SRT critical value of 10 days. Removal of carbamazepine, a notoriously recalcitrant PPCP, is not effected by SRT and does not have a SRT.

Both biodegradation and adsorption are highly dependent on both hydraulic and sludge retention times (HRT and SRT). HRT differs from SRT and is a measurement of the average length of time wastewater remains in a treatment system. Miège et al., 2008 found that treatment plants that have a high SRT (>12 days) and a high HRT (>10 days) are better at removing CECs than those with low SRT and HRT. The longer the wastewater is able to interact with the sludge, the more biodegradation and adsorption can occur for some CECs.

[5] (Hoque et al., 2014; X. Li et al., 2013), summer removal rates reported

This statement holds for all treatment technologies that rely on biodegradation for treatment. Advantages and disadvantages of CAS as a CEC and nutrient treatment technology are highlighted in Table 4.

**Table 4. Conventional activated sludge advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[6] |
|---|---|---|---|
| • Smaller footprint than natural treatment system | • Does not remove recalcitrant CECs<br>• Highly variable removal rates depending on local conditions | • Capable of limited nutrient removal<br>• Cannot get nutrient concentration low enough for some receiving waters | Caffeine- 99.3%<br>Ibuprofen- 93.5%<br>Triclosan- 34-99%<br>Carbamazepine- >0% |

## Attached Growth Systems

Attached growth refers to systems where microorganisms attach to media and form a biofilm layer where biodegradation, oxidation, adsorption, and absorption of CECs can occur. This includes trickling filters and integrated fixed-film activated sludge systems. Moving bed biofilm reactors are technically an attached growth system but are discussed separately in this paper (see sub-section below) due to their enhanced ability to remove CECs and denitrify.

Attached growth systems have potential to be effective at removing CECs. The biofilm used can promote the growth of slow growing material and expand the number of active microbes in the system. This allows for more CECs to be removed even with a shorter SRT than CAS (Grandclément et al., 2017). However, like other systems mentioned, removal rates are highly dependent on local conditions. Studies show inconsistent removal rates of CECs between seasons and WWTPs. Advantages and disadvantages of attached growth systems as a CEC and nutrient treatment technology are highlighted in Table 5.

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[6] (Blair et al., 2015; Krzeminski et al., 2019; Sipma et al., 2010)

**Table 5. Attached growth systems advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[7] |
|---|---|---|---|
| • Relatively inexpensive to maintain and operate | • Removal rates are highly dependent on characteristics of the wastewater and type of CEC<br>• Variable removal rates | • Similar to conventional activated sludge | Caffeine- N/A<br>Ibuprofen- N/A<br>Triclosan- N/A<br>Carbamazepine- N/A |

## Moving Bed Biofilm Reactors (MBBR)

MBBRs are a type of attached growth system of particular interest due to their ability to denitrify and be added as either a tertiary step (Kermani et al., 2008) or as a retrofit to an existing aerobic tank. Recent studies that indicate that MBBR is more efficient at removing CECs than other attached growth systems or CAS (Krzeminski et al., 2019). Removal of pharmaceuticals are usually co-metabolized with dissolved organic carbon and nitrogen.

Casas et al., 2015 studied an MBBR system used as pretreatment for hospital wastewater. They found that 21 of the 26 pharmaceuticals they monitored for were removed by at least 20%. MBBR shows promise as a potential pretreatment wastewater technology.

Additionally, MBBR systems can be optimized by changing the media material and operational conditions to enhance removal rates of specific CECs or for CECs overall (Krzeminski et al., 2019). Advantages and disadvantages of MBBR as a CEC and nutrient treatment technology are highlighted in Table 6.

[7] No published removal rates are provided because most studies reported removal rates by CECs type (e.g. pharmaceuticals, hormones, etc.).

**Table 6. Moving bed biofilm reactors advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[8] |
|---|---|---|---|
| • Can be easily installed in retrofitted activated sludge tanks<br>• Higher SRT leads to a greater CEC removal than other attached growths systems and CAS | • CEC removal is dependent on local conditions<br>• Variable removal rates | • High removal of nutrients under ideal conditions | Caffeine- 99%<br>Ibuprofen- 93%<br>Triclosan- 80-92%<br>Carbamazepine-<br>0-75% |

## Membrane Bioreactor (MBR)

Recent studies show that MBR can be effective in removing pharmaceutical and personal care products and other CECs that are otherwise resistant to removal in trickling filters and activated sludge treatment processes. The MBR process retains all the removal capabilities of an activated sludge system with the added benefit of a filter, eliminating the need for secondary clarification. MBR has a much longer sludge retention time (SRT) than conventional activated sludge and it is believed that the main removal pathway for CECs is through adsorption (Tadkaew et al., 2011).

Adsorption occurs usually at the membrane filter where a thin sludge film forms. Adsorption does not occur for hydrophilic CECs, but slightly hydrophilic and hydrophobic compounds are able to adsorb during the MBR process. MBR systems are more effective at removing CECs than an activated sludge system, due to a higher SRT leading to better biodegradability (Das et al., 2012). For example, Le-Minh et al., 2010 found that MBR had 75-95% removal rates for antibiotics compared to 0-66% removal rates when using conventional activated sludge (CAS).

Additionally, multiple MBRs can be put in sequence or paired with other technologies (e.g., chemical addition or biological selectors) which has the potential to increase the efficacy of both nutrient and CEC removal. De La Torre et al., 2015 found that integrated fixed film activated sludge (IFAS) combined with MBR was more effective at removing pharmaceuticals compared to MBR and CAS. C. Li et al., 2015 found that MBR in conjunction with granular and powdered activated carbon (GAC and PAC respectively) was able to remove over 90% of the carbamazepine present in the influent. Advantages and disadvantages of MBR as a CEC and nutrient treatment technology are highlighted in Table 7.

[8] (Krzeminski et al., 2019; Luo et al., 2014)

**Table 7. Membrane bioreactor advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[9] |
|---|---|---|---|
| • Retains all the removal capabilities of an activated sludge system with the added benefit of a filter<br>• More effective than CAS<br>• Small footprint | • Not as effective at removing hydrophilic CECs<br>• Membrane fouling can occur<br>• High energy costs during operation | • Better nutrient removal than CAS –requires additional treatment steps | Caffeine- 100%<br>Ibuprofen-100%<br>Triclosan- 99%<br>Carbamazepine- 28% |

# Biological Nutrient Removal

Biological Nutrient Removal (BNR) is a widely used enhancement to secondary treatment. This technology removes more nutrients than conventional activated sludge. BNR treatment configurations differ depending on whether removing nitrogen or phosphorus. BNR encourages the growth of different microorganisms by creating basins with different conditions, namely aerobic, anaerobic, and anoxic. Having multiple basins increases the HRT and SRT, which also encourages a more diverse microbial community. Having multiple basins also increases HRT, and in multiple studies, BNR removed more pharmaceuticals and other CECs than conventional activated sludge, attached growth systems, and membrane bioreactors (S. Kim et al., 2018; Okuda et al., 2008; Smook et al., 2008).

Okuda et al., 2008 concluded that Conventional Activated Sludge had 1.5 times higher concentrations of pharmaceuticals in its effluent compared to effluent from a facility with BNR. This is because BNR adds extra stages to the wastewater treatment process, which increases the solids retention time (SRT).

Increasing SRT is considered the important parameter affecting the removal of CECs from wastewater with activated sludge (Sipma et al., 2010). A longer SRT increases the likelihood that adsorption of degradation of CECs can take place, as described in the CAS section. Advantages and disadvantages of BNR as a CEC and nutrient treatment technology are highlighted in Table 8.

[9] (M. Kim et al., 2014; Krzeminski et al., 2019; Sipma et al., 2010)

**Table 8. Biological nutrient removal advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates [10] |
|---|---|---|---|
| • Increased HRT and SRT allow for more CEC removal than CAS | • Highly variable removal rates<br>• Does not remove recalcitrant compounds | • Technology designed specifically for nutrient removal | Caffeine- 80%<br>Ibuprofen- 87%<br>Triclosan- 69%<br>Carbamazepine-<30% |

## Carbon Filtration

Carbon filtration is able to remove many recalcitrant CECs from water. Granular activated carbon (GAC) and powdered activated carbon (PAC) are the most commonly used forms of carbon filtration available. Activated carbon particles are porous and therefore have a larger surface area for CECs to sorb onto (Rajapaksha et al., 2019). Many different types of CECs are able to sorb onto activated carbon and it does not produce toxic byproducts like some removal technologies. This treatment technology can only be used in conjunction with others as a part of a treatment train (Fig. 5). It is often used after advanced oxidations to remove the toxic byproducts produced (M. B. Ahmed et al., 2017). Removal rates are highly dependent on the type of activated carbon used and companies report variable removal rates between different products.

Currently, activated carbon can be expensive to use, maintain, and dispose of as spent materials. Activated carbon needs to continually be replaced or regenerated to ensure efficient sorption of CECs (Wang & Wang, 2016). Additionally, the complex nature of wastewater could have matrix effects on removal rates of CECs using carbon filtrations (Kyzas et al., 2015). Advantages and disadvantages of carbon filtration as a CEC and nutrient treatment technology are highlighted in Table 9.

[10] (Okuda et al., 2008; Yu et al., 2006), Carbamazepine removal rate is from a different study than caffeine, ibuprofen and triclosan.

**Table 1. Carbon filtration advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[11] |
|---|---|---|---|
| • Removes recalcitrant CECs, including PFAS | • Expensive to set up, run, and operate<br>• Creates spent materials needing disposal<br>• Usually added on to a treatment train and not a standalone treatment | • Minimal<br>• Technology not intended to remove nutrients | Caffeine-100%<br>Ibuprofen- 99%<br>Triclosan- 95%<br>Carbamazepine- 99% |

## Addition of Biosorbants

As noted, activated carbon can be expensive and needs to be periodically regenerated or replaced. Scientists are looking into more cost effective alternatives to activated carbon with the addition of biosorbants that would increase sorption of CECs and increased microbial degradation. These include agricultural wastes, sterilized sludge, and marine algae (M. B. Ahmed et al., 2017; Rajapaksha et al., 2019).

Agricultural waste like rice straw, saw dust, and spent mushroom material is inexpensive and abundant. These materials can act similarly to activated carbon, providing adsorption sites. Most of these studies are in the pilot phase and have not been scaled up for use in a WWTP. However, biosorbants have shown promising results and many were able to remove the most recalcitrant medication (Costa et al., 2019). Most notably, the addition of rice straw removed 60-75% of carbamazepine, a pharmaceutical that is notoriously difficult to remove, (Liu et al., 2013). Advantages and disadvantages of the addition of biosorbants as a CEC and nutrient treatment technology are highlighted in Table 10.

[11] (Luo et al., 2014)

**Table 10. Biosorbants advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates |
|---|---|---|---|
| • Can be added to most existing treatment technologies<br>• Biosorbants can be local, inexpensive bioproducts | • Highly variable removal rates<br>• Increase in biosolids production with potential CEC contamination<br>• Requires trial and error<br>• Newer, unproven technology | • Highly dependent on biosorbant and local conditions | • Highly dependent on the biosorbants used<br>• Rice straw biosorbant was able to remove 60-75% of carbamazepine in lab studies |

## Advanced Oxidation Processes

Advanced oxidation processes (AOPs) have shown the most promise in reducing all CECs in wastewater. AOP can destroy CECs that do not sorb easily or are only partially removed during other processes. Many WWTPs currently use UV light as a final disinfecting stage and can help remove some CECs. While UV light alone is able to destroy microorganisms in wastewater, it is not a strong enough oxidant to successfully breakdown many CECs in wastewater.

UV light can be combined with hydrogen peroxide or titanium dioxide to create a stronger oxidant. These methods have both been used to reduce CEC loading in wastewater and has been demonstrated to remove some recalcitrant CECs (Anupama & Shrihari, 2018; Dai et al., 2012; Rosenfeldt & Linden, 2004). However, removal rates can be variable. Not enough research has been done to fully understand how chemical matrices, pH, and temperature effect removal rates for every CEC (Wang & Wang, 2016). Moreover, oxidation processes like UV can break down CECs into more harmful byproducts (Park et al., 2016; Wert et al., 2007). Advantages and disadvantages of AOP as a CEC and nutrient treatment technology are highlighted in Table 11.

**Table 11. Advanced oxidation process advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[12] |
|---|---|---|---|
| • Basic UV disinfection already installed in many WWTP plants as final disinfection step<br>• Often already installed in WWTP plants as final disinfection step | • Causes toxic byproducts<br>• Does not break down all CECs compared to other AO processes<br>• Takes longer than ozonation | • None<br>• Technology not intended to remove nutrients | Caffeine- N/A<br>Ibuprofen- 34%<br>Triclosan- N/A<br>Carbamazepine- 23% |

## Ozonation

Ozonation is currently the most commonly used method for removal of CECs from wastewater. Ozone gas is generated onsite and infused into wastewater, usually as an ultimate and penultimate step in the treatment process. Ozonation relies on the creation of hydroxyl radicals ($OH^-$). Hydroxyl radicals react with CECs to cleave bonds and thus destroy the CEC. More stable CECs require stronger oxidants, like ozone to destroy them. Ozonation can remove most PPCPs by 90% (Wang & Wang, 2016). Advantages and disadvantages of ozonation as a CEC and nutrient treatment technology are highlighted in Table 12.

**Table 12. Ozonation advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[13] |
|---|---|---|---|
| • Stronger oxidant than UV that can break down all known CECs | • Can cause toxic byproducts<br>• Expensive to install and run<br>• Energy intensive | • None<br>• Technology not designed to remove nutrients | Caffeine- 100%<br>Ibuprofen- 83%<br>Triclosan- 99%<br>Carbamazepine- 100% |

[12] (Luo et al., 2014) (Anupama & Shrihari, 2018), removal rates are after 10 minutes of UV radiation. Removal rates vary greatly with time and wavelength.
[13] (Chopra & Kumar, 2020, Luo et al., 2014, Wang & Wang, 2016)

## Microfiltration and Ultrafiltration

WWTPs mainly use microfiltration or ultrafiltration after primary and secondary treatment to remove particulate phosphorous. The pore size of these filters is much larger than aqueous CECs, so the filter itself is ineffective at removing most CECs. Filters are able to remove hydrophobic CECs but do not remove hydrophilic compounds well (S. Kim et al., 2018). Additionally, the chemistry of the source water effects removal rate of CECs making it harder to predict and guarantee removal rates with this technology. Micro- and ultrafiltration are often used in a membrane bioreactor system. Advantages and disadvantages of microfiltration and ultrafiltration as CEC and nutrient treatment technologies are highlighted in Table 13.

**Table 23. Microfiltration and ultrafiltration advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[14] |
|---|---|---|---|
| • Removes CECs with a higher molecular weight | • Requires energy to run<br>• Membrane fouling can occur<br>• Removal rate dependent on physical conditions of the wastewater | • Nutrient removal is dependent on local conditions (pH, Temp, etc.)<br>• Nutrient removal requires enhanced coagulation through chemical addition ahead of filtration | Caffeine- <70%<br>Ibuprofen- <70%<br>Triclosan- <70%<br>Carbamazepine- 0% |

## Nanofiltration

Nanofiltration (NF) is a newer technology that tends to be more expensive than micro- and ultrafiltration. The pore size is smaller than micro- and ultrafiltration systems and requires higher pressure to pass untreated water through. The smaller pore size filters out CECs and other substances on the molecular scale, whereas micro and ultrafiltration are only able to filter out macromolecules like colloids and proteins. For example, NF is able to remove 80-100% of carbamazepine which is notoriously recalcitrant in wastewater (Luo et al., 2014). NF does not rely on a biofilm forming on the filter to remove CECs (Patel et al., 2019). Advantages and disadvantages of NF as a CEC and nutrient treatment technology are highlighted in Table 14.

[14] (S. Kim et al., 2018)

**Table 14. Nanofiltration advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates[15] |
|---|---|---|---|
| • Effective at removing most CECs | • Requires a lot of energy and expertise to maintain<br>• Increased cost<br>• Membrane fouling can occur<br>• Removal of CEC also dependent on molecular size of CEC<br>• Doesn't remove all CECs | • High level of phosphorous and nitrate removal due to molecular size | Caffeine- 68%<br>Ibuprofen- 30-50%<br>Triclosan- 40-60%<br>Carbamazepine- 80-100% |

## Reverse Osmosis

Reverse Osmosis (RO) works by using pressure to pass wastewater through a porous filter in the opposite direction of natural osmosis. Like the other filtration processes, removal of CECs through RO depends on membrane properties and CEC characteristics. RO uses a filter with the smallest pore size of any filtration method and has long been used for drinking water purposes. CEC removal is high, and RO can even remove solutes that are resistant to any other WWTP technology. This includes removing 95% of carbamazepine and PFAS (M. Kim et al., 2014; Patel et al., 2019).

RO is considered one of the most effective treatments for removing CECs. However, it is expensive to install and run, and has a lower maximum output compared to other filtration processes. Additionally, RO can be energy intensive because it requires pressure to overcome the natural direction of osmosis and generates a concentrated waste brine that requires disposal. Advantages and disadvantages of RO as a CEC and nutrient treatment technology are highlighted in Table 15.

[15] (S. Kim et al., 2018, Luo et al., 2014)

**Table 15. Reverse osmosis advantages, disadvantages, nutrient removal capabilities, and CEC removal rates**

| Advantages | Disadvantages | Nutrient Removal Capabilities | Published CEC Removal Rates [16] |
|---|---|---|---|
| • Nearly complete removal of CECs | • High upfront costs<br>• Requires a lot of energy to run<br>• Membrane fouling can occur<br>• Need skilled personnel to run<br>• Disposal needed for waste brine | • High level of nutrient removal | Caffeine- 99%<br>Ibuprofen- 99.4%<br>Triclosan- 94%<br>Carbamazepine- 99% |

# Conclusions

Wastewater can contain pharmaceuticals, personal care products (e.g. synthetic fragrances, antibacterial compounds), plasticizers, food additives, flame retardants, microparticles, and per- and polyflouryl alkyl substances (PFAS) which can have a negative impact on aquatic health of receiving waters. Adding more advanced treatment to a wastewater treatment plant can reduce some or most CECs, with the removal rate dependent upon the approach chosen and other implementation factors. For most CECs, concentrations may be reduced but the chemicals will not be not entirely removed from WWTP effluent and the pollutant loadings could be substantial. Some CECs are so recalcitrant that they will continue to remain in the effluent unless the most advanced (and expensive) approaches are selected. More work remains to understand the impact of CEC loadings and their effects on human health and aquatic life.

This paper reviewed fifteen categories of treatment technologies, and their observed treatment effectiveness for four indicator CECs. In summary:

- **Caffeine** was successfully removed (90-100%) by nearly all of the technologies investigated in this review. Microfiltration and ultrafiltration, and nanofiltration were the least effective approaches with removal rates under 70%.
- **Carbamazepine** was very successfully removed (99%) by carbon filtration, ozonation, and reverse osmosis. Nanofiltration removed 80-100%, and moving bed biofilm reactors removed between 0-75%. The other technologies all reduced concentrations by <25%, or not at all.

[16] (S. Kim et al., 2018; Rahman et al., 2018)

- **Triclosan** was successfully removed (90-99%) by carbon filtration, membrane bioreactors, UV advanced oxydation, ozonation, and reverse osmosis. Onsite sewage systems removed 80%; moving bed biofilm reactors removed 80-100%; constructed wetlands removed 58-79%; and conventional activated sludge removed 34-99%.
- **Ibuprofen** was removed (>75%) by all technologies except for lagoons, UV advanced oxidation, microfiltration and ultrafiltration, and nanofiltration.

Overall, carbon filtration, ozonation, and reverse osmosis were the most successful CEC removal technologies across the four indicator CECs. Among these three tertiary technologies, only reverse osmosis successfully removes nutrients. This is due to molecular level physical separation through the semi-permeable membrane requiring large amounts of energy. Wastewater design engineers may want to focus on other more effective advanced treatment technologies when designing treatment upgrades solely for nutrient removal.

Among nutrient removal technologies evaluated, moving bed biofilm is the most promising at removing CECs. The nutrient removal technology with the second highest rate of CEC removal is membrane bioreactors. Membrane bioreactors performed well for all indicator CECs except carbamazepine, which is a notoriously recalcitrant CEC. Biological nutrient removal and activated growth systems have the lowest rate of CEC removal among the nutrient removal technologies evaluated. BNR and activated growth systems also remove caffeine and ibuprofen but not carbamazepine. BNR and activated growth systems were not evaluated for triclosan removal.

Conventional activated sludge, constructed wetlands, and onsite systems also perform reasonably well in removing CECs that adsorb well. This study also found that most filtration systems are effective in removing CECs, with those with the smallest pore size working the best. Biosorbant is a promising treatment technology, however removal rates are highly dependent on the product used.

It is currently not known whether the level of CEC reduction expected from the most accessible WWTP technologies will be adequate to reduce or prevent the harmful effects of CECs on aquatic life or if reduction is even necessary for all CECs found in wastewater. The reasons for this uncertainty are threefold:

1. There currently is not enough information about both acute and chronic toxicity of CECs and their degradants on Pacific Northwest aquatic life.
2. It is unclear if the concentrations of all CECs found in wastewater is toxic at the concentrations observed in wastewater studies in the region. There is not enough information on the chronic toxicity of most of the CECs found in wastewater on different aquatic species and throughout their lifecycles. Moreover, there is no information about the effect of a matrix of CECs on aquatic health.
3. Many of the most toxic CECs are also the most recalcitrant and will remain in wastewater even with more advanced treatment options.

Fully addressing CECs in wastewater will require more than just upgrading WWTP technologies. Recommendations to further address CECs in wastewater are outlined in the next section.

# Recommendations

The recommendations below were based on the following sources:

- The Association of Clean Water Administrators and Association of Safe Drinking Water Administrators (Association of Clean Water Administrators & Association of State Drinking Water Administrators, 2019)
- Water Europe (Joint Norman & Water Europe Position Paper, 2019)
- The EPA (Harding et al., 2006), California Waterboards (Phonsiri et al., 2019); and from
- Recommendations included in the peer-reviewed literature and previous Ecology reports discussed earlier in this document; and considering further input from Department of Ecology staff.

1. Develop prioritized lists of CECs to track and study further in Washington State

   Washington should focus on the CECs that are known to be in the State's waters at concentrations that may affect human health and aquatic biota, and should create a prioritized list of the most deleterious CECs. This prioritized list should be reassessed occasionally using an established method for adding and removing CECs from the list as they are introduced, studied, and addressed following approaches by other agencies that have done this.

   California has a list of prioritized chemicals for the San Francisco Bay Estuary (Sutton et al., 2013); the Association of Clean Water Administrators and the Association of Safe Drinking Water Administrators also call for a national prioritized list of CECs (Association of Clean Water Administrators & Association of State Drinking Water Administrators, 2019). Scientists are currently developing a Washington-specific list as of summer 2020.

2. Study Prioritized CECs Discharges in WWTP Effluents across Washington State
   - Determine discharge concentrations and loads of prioritized CECs (from recommendation 1) from WWTPs in Washington.
     - Ecology should conduct or support a large study, using results from the Washington-specific prioritization list, to provide a better understanding of the occurrence and magnitude of CECs discharged from WWTPs in Washington State. Ecology, or WWTPs, should also conduct a study of newly installed nutrient removal technologies to determine the extent to which they are also removing CECs.

3. Develop a Washington-specific list of indicator CECs and approved testing methods.

   A broadly accepted list of indicator CECs with approved testing methods would allow WWTPs to get a better understanding of their CEC removal rates and overall efficacy without having to sample influent and effluent for all known CECs.

   Removal rates are highly variable and effected by local conditions at WWTPs. Testing for all known CECs is expensive, impractical and sometimes impossible because there are no approved methods. The indicator approach would allow for comparison of removal rates among WWTPs and across the state. The EPA (Kostich et al., 2014) and other environmental governing bodies, including the State of California (Anderson et al., 2010) and several countries in Europe (Joint Norman & Water Europe Position Paper, 2019), have developed lists of indicator compounds that help determine removal efficiencies and monitoring of CECs.

4. Assist WWTPs in upgrading and optimizing facilities

   Ecology should support WWTPs that may need to upgrade their systems to reduce nutrients and choose technologies that can also reduce CEC discharges. This paper as well as a past Ecology paper (Lubliner et al., 2010) shows there is potential to remove some CECs by upgrading WWTPs to include technologies or adjust plant operations in ways that increase hydraulic and/or sludge retention times. Coordinating with planned upgrades for nutrient removal and anticipated plant expansions can be a cost effective way to achieve these multiple goals. It is important to account for some potential negative environmental impacts like increased energy usage and the need for other non-renewable products for treatment.

5. Increase education and outreach about proper disposal of unused pharmaceuticals

   While there is little to be done about pharmaceuticals entering WWTPs through excretion, it is possible to limit the amount of medications directly flushed. There is conflicting guidance on how to dispose of medication from the Federal Drug Administration (FDA), EPA, and Ecology. The Washington State Department of Health manages the Safe Medication Return Program. This program is designed to reduce abuse, fatal overdoses, and poisoning caused by unused prescription medicines. However, take back programs can have an ancillary benefit of ensuring prescription drugs do not get flushed.

   Below are three groups that routinely dispose of pharmaceuticals and prescription drugs. Increased outreach and education around this issue can help reduce the amount of pharmaceuticals that are improperly disposed into the sewer system:

#### Household

The Federal Food and Drug Administration recommends flushing pharmaceuticals as a disposal method if a take back program is not available. [17] Furthermore, the FDA recommends immediately flushing unused opioids. [18] However, this is not considered best practice when considering WWTPs. The EPA and Washington State Law (RCW 69.48) do not support flushing medications for disposal unless a patient or care provider is specifically instructed to do so.[19] While there are drug take back programs set up in many communities across the state, a lot of people find it more convenient to simply flush the medication, not realizing the eventual impact to receiving waters. A consistent, widespread education campaign can help curtail flushing of medications by the general public. The Department of Health launched the Safe Medication Return Program in November 2020 which can address this issue.

### Animal Facilities

Veterinarians also use and prescribe many of the same pharmaceuticals as humans. Farmers often give hormones, antibiotics, and other medication to their animals. The American Veterinarian Medical Association has guidance that is in line with Washington State regulation (RCW 69.48).[20] Increased education and information on proper disposal for pet owners, livestock handlers, and veterinarians can assure compliance with these recommendations and regulations.

### Medical Facilities

Medical facilities can be large sources of pharmaceuticals and other CECs because they generate a lot medical waste. Ecology developed a Pharmaceutical Waste Disposal Guide for healthcare facilities that details how medication should be disposed. Increased education and information on proper disposal can assure compliance with this guide.

6. Continued Research
   - Emerging Technologies

     Wastewater treatment technologies are constantly evolving, especially with regard to CEC removal and our collective understanding of their efficacy. There are many emerging technologies that could be inexpensive and effective to use but lack the rigorous scientific review at the moment (i.e. biosorbants). Ecology should continue to review the available science for potential CEC removal technologies.

   - Improve Understanding of Ambient Conditions, Aquatic Health, and Human Health

[17] https://www.fda.gov/consumers/consumer-updates/where-and-how-dispose-unused-medicines
[18] https://www.fda.gov/media/109643/download
[19] For more information visit EPA's website on drug disposal and Washington Department of Health's website.
[20] American Veterinary Medical Association guidance is available here

Washington has a limited understanding of the extent, type and amount of CECs in the state's ambient water. Washington has 600 individually permitted WWTPs across the state and only four of them have limited information about CEC concentrations in their effluent. A more extensive ambient groundwater and surface water monitoring program should include CEC analysis across Washington State. Coupled with the prioritized list from Recommendation 1, Ecology can then address CECs of known concern and their effects on aquatic health.

Developing toxicity data is an expensive and long process that Ecology does not have the resources to repeatedly do. Because the information is of national relevance, Ecology encourages the EPA to conduct further research on the toxicity of CECs in wastewater, and the synergistic or matrix effect of CECs on aquatic life. EPA is better able to continue to stay apprised of new toxicity data as it emerges from other sources. This information can inform the level of removal that is needed to maintain the aquatic health of receiving water or be used to develop human health and aquatic life criteria.

- Improve Understanding of CECs in Stormwater and Agricultural Runoff

  CECs in urban stormwater and other land surface runoff are outside the scope of this paper, but more information is needed to fully understand the potential pathways and impact of pharmaceuticals and personal care products in stormwater and inform regional strategies for addressing them.

  Although it is not the primary pathway for these CECs to reach large receiving waters, examples of urban stormwater runoff containing pharmaceuticals and personal care products, including methamphetamines and ibuprofen, have been documented (Peter et al., 2020), and agricultural runoff from areas where livestock are treated with medications is another possible source of pharmaceuticals.

  Runoff should therefore be included as a pathway for consideration when seeking a more holistic understanding of these chemicals' impacts on biota. Broader sampling of indicator CECs across the urban and agricultural landscape could inform areas where this problem is most likely to occur. There is a lot of overlap in combined sewer overflow treatment and wastewater treatment processes, and likely their efficacy in removing CECs.

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# Appendix A. Acronyms and Abbreviations

| | |
|---|---|
| **AOP** | Advanced Oxidation Process |
| **BNR** | Biological Nutrient Removal |
| **CAS** | Conventional Activated Sludge |
| **CEC** | Contaminants of Emerging Concern |
| **CWA** | Clean Water Act |
| **DEHP** | Di(2-ethylhexyl)phthalate |
| **DEP** | Diethyl phthalate |
| **DIOP** | 2,3-*O*-isopropylidene-2,3-dihydroxy-1,4- |
| **EPA** | Environmental Protection Agency |
| **FOEN** | Swiss Federal Office on the Environment |
| **FWS** | Free Water Surface |
| **GAC** | Granular Activated Carbon |
| **GEA** | German Environmental Agency |
| **HRT** | Hydraulic Retention Time |
| **MBBR** | Moving Bed Biofilm Reactors |
| **MBR** | Membrane Bioreactor |
| **MF** | Microfiltration |
| **NF** | Nanofiltration |
| **NPDES** | National Pollutant Discharge Elimination System |
| **OH-** | Hydroxyl radical |
| **PAC** | Powdered Activated Carbon |
| **PBDE** | Polybrominated diphenyl ethers |
| **PFAS** | Per and polyflourylalkyl substances |
| **PFBA** | Pentafluorobenzoic acid |
| **PFHxA** | Perfluorohexanoic acid |
| **PFOA** | Perfluorooctanoic Acid |
| **PFPeA** | Perfluoropentanoic Acid |
| **PPCPs** | Pharmaceuticals and Personal Care Products |
| **PSP** | Puget Sound Partnership |
| **RO** | Reverse Osmosis |
| **SFW** | Subsurface Flow Wetlands |
| **SRKW** | Southern Resident Killer Whale |
| **SRT** | Sludge Retention Time |
| **SWAM** | Swedish Agency for Marine and Water Management |
| **UF** | Ultrafiltration |
| **WWTP** | Wastewater Treatment Plant |

# Appendix B. Summary of CEC Removal Capabilities

| Technology | Advantages | Disadvantages | Nutrient removal capabilities | Published CEC removal rates (%) | | | |
|---|---|---|---|---|---|---|---|
| | | | | Caffeine | Carbamazepine | Triclosan | Ibuprofen |
| **Onsite Sewage Systems** | • CEC removal rates on par with conventional activated sludge<br>• Good for remote areas without access to larger facilities | • Does not remove recalcitrant compounds<br>• Not easily upgraded<br>• Removal rates are highly variable based local conditions<br>• Can fail | • Some nutrient removal does occur<br>• Can also be a source for nutrients when drain field fails or is poorly designed | 99.63 | 8 | 80 | 76 |
| **Constructed Wetlands** | • Long HRT increases removal rates<br>• More mechanisms for removal than on-site sewage systems<br>• Good for remote areas without access to larger facilities | • Requires space<br>• Does not remove recalcitrant CECs<br>• Removal rate is susceptible to changing environmental conditions | • Higher nutrient removal rates than lagoons and on-site sewage systems on average | 80.3 | 26.7 | 58-79 | 80 |
| **Lagoons** | • Long retention time increases removal rates<br>• Inexpensive and easily maintained<br>• Good for remote areas without access to larger facilities | • Does not treat recalcitrant CECs that do not easily adsorb<br>• Variable removal rates depending on local conditions<br>• Requires large land area, which can increase the cost | • Frequently considered a source for ammonia | 100 | 0 | 97.2 | 91.5 |

| Technology | Advantages | Disadvantages | Nutrient removal capabilities | Published CEC removal rates (%) | | | |
|---|---|---|---|---|---|---|---|
| | | | | Caffeine | Carbamazepine | Triclosan | Ibuprofen |
| **Conventional activated sludge (CAS)** | • Smaller footprint than natural treatment system | • Does not remove recalcitrant CECs<br>• Highly variable removal rates depending on local conditions | • Capable of removing nutrients<br>• Cannot get nutrient concentration low enough for some water bodies | 99.3 | 0 | 34-99 | 93 |
| **Attached growth systems** | • Relatively inexpensive to maintain and operate | • Removal rates are highly dependent on characteristics of the wastewater and type of CEC<br>• Variable removal rates | • Similar to conventional activated sludge | N/A | N/A | N/A | N/A |
| **Moving bed biofilm reactors** | • Can be easily installed in retrofitted activated sludge tanks<br>• Higher sludge retention time leads to greater CEC removal | • CEC removal is dependent on local conditions<br>• Variable removal rates | • High removal of nutrients under ideal conditions | 99 | 0-75 | 80-92 | 93 |
| **Membrane bioreactors** | • Retains all the removal capabilities of an activated sludge system with the added benefit of a filter<br>• More effective than CAS<br>• Small footprint | • Not as effective at removing hydrophilic CECs<br>• Membrane fouling can occur<br>• More energy costs when running it | • Better nutrient removal than CAS | 100 | 28 | 99 | 100 |

| Technology | Advantages | Disadvantages | Nutrient removal capabilities | Published CEC removal rates (%) | | | |
|---|---|---|---|---|---|---|---|
| | | | | Caffeine | Carbamazepine | Triclosan | Ibuprofen |
| **Biological nutrient removal** | • Increased HRT and SRT allow for more CEC removal | • Highly variable removal rates<br>• Does not remove recalcitrant compounds | • Good; the technology is designed specifically for nutrient removal | 80 | <30 | 69 | 87 |
| **Carbon filtration** | • Removes recalcitrant CECs, including PFAS | • Expensive<br>• Creates spent materials<br>• Not a standalone treatment | • Minimal; the technology not intended to remove nutrients | 100 | 99 | 95 | 99 |
| **Biosorbants** | • Can be added to most existing treatment technologies<br>• Biosorbants can be local, inexpensive byproducts | • Highly variable removal rates<br>• Increase in biosolids production with potential CEC contamination<br>• Requires trial and error | • Highly dependent on biosorbant and local conditions | Highly dependent on the biosorbants used | | | |
| **UV Advanced oxidation** | • Often already installed in WWTP plants as final disinfection step | • Results in disinfection byproducts and degradation byproducts<br>• Does not break down all CECs<br>• Takes longer than ozonation | • None; the technology is not intended to remove nutrients | N/A | 23 | N/A | 34 |
| **Ozonation** | • Stronger oxidant that can break down all known CECs | • Can cause disinfection byproducts<br>• Expensive<br>• Energy intensive | • None<br>• Technology not designed to remove nutrients | 100 | 100 | 99 | 83 |

| Technology | Advantages | Disadvantages | Nutrient removal capabilities | Published CEC removal rates (%) | | | |
|---|---|---|---|---|---|---|---|
| | | | | Caffeine | Carbamazepine | Triclosan | Ibuprofen |
| **Microfiltration and ultrafiltration** | • Removes CECs with a higher molecular weight | • Requires energy to run<br>• Membrane fouling can occur<br>• Removal rate dependent on physical conditions of the wastewater | • Nutrient removal is condition dependent<br>• Nutrient removal requires enhanced coagulation beforehand | <70 | 0 | <70 | <70 |
| **Nanofiltration** | • Effective at removing most CECs | • Requires a lot of energy and expertise to maintain<br>• Increased cost<br>• Membrane fouling can occur<br>• Does not removal all CECs; removal depends on molecular size | • High level of phosphorous and nitrate removal due to molecular size | 68 | 80-100 | 40-60 | 30-50 |
| **Reverse osmosis** | • Nearly complete removal of CECs | • High upfront costs<br>• High energy use<br>• Membrane fouling can occur<br>• Requires skilled personnel<br>• Need to dispose of brine | • High level of nutrient removal | 99 | 99 | 94 | 99.4 |

Advantages, disadvantages, nutrient removal efficiency, and reported removal efficiencies by technologies used in wastewater treatment for four common compounds: three pharmaceuticals and one personal care product. (See Tables 2-16 for references.)